

# PROCESS FOR FORMING ALLOYS IN SITU IN ABSENCE OF LIQUID-PHASE SINTERING

## FIELD OF THE INVENTION

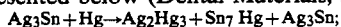
The invention relates to a process of forming intermetallic alloys. In a preferred embodiment, the invention relates to processes for forming the intermetallic alloys as in situ dental restorations.

## BACKGROUNDS

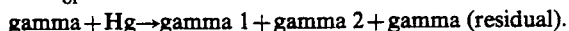
Amalgams and related alloys have been incorporated into a variety of commercial applications and thus a number of processes for producing such amalgams are known. For example, U.S. Pat. No. 4,664,855 discloses a universally employed process which triturates elemental metallics or an intermetallic alloy, in the form of comminuted filings or atomized spherical powders, with the sintering agent mercury and compacts the resulting amalgam into a uniform, consolidated intermetallic alloy. The process may be considered a combination of liquid phase and reactive metal sintering. The finely comminuted metallic or intermetallic powders react with the Hg and when pressure is applied to the reaction product, form a compact, high density mass. U.S. Pat. No. 3,933,961 discloses a process for preparing a pre-weighed alloy tablet of uniform weight which is then triturated with a weighed quantity of Hg to form a traditional amalgam alloy.

Metallic powders have two basic forms: 1) minute lathe-cut filings and 2) atomized, spherical particles. Lathe-cut filings are subsequently milled and sifted to produce the desired particle size. The length of particles in a commercial lathe-cut alloy might range from 60-120  $\mu\text{m}$  long, 10-70  $\mu\text{m}$  wide and from 10-35  $\mu\text{m}$  thick. On the other hand, spherical particles, produced by means of an atomizing process whereby a spray of tiny drops is allowed to solidify in an inert gaseous (e.g., argon) or liquid (e.g., water) environment, have a maximum size between 40 and 50  $\mu\text{m}$  (Dental Amalgams, William J. O'Brien, Ph.D., Quintessence Publishing Co., 1989, at p. 264, 266).

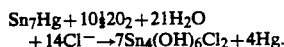
For more than 150 years, dental practitioners have applied a Ag-Sn system and related intermetallic alloys as dental amalgam preparations for use in dental restorations such as fillings and prostheses. The amalgamation reaction which forms Ag-Sn dental preparations is best represented below (Dental Materials, supra, at p. 269);



or



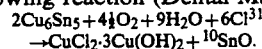
After completion of the amalgamation reaction, the high-melting  $\text{Ag}_3\text{Sn}$  (gamma) products are embedded in a matrix of mercury reaction products. Both the  $\text{Ag}_2\text{Hg}_3$  (gamma 1) and  $\text{Sn}_7\text{Hg}$  (gamma 2) phases form a continuous network. In the amalgam structure above, however, the  $\text{Sn}_7\text{Hg}$  is susceptible to corrosion by the following reaction:



Dental amalgam corrosion results in a weakened structure and further formation of unsupported, "margin" amalgam which easily fractures under tension.

The introduction of contemporary amalgams in the 1960s (amalgams known as "high-copper" amalgams) eliminated the corrosive phenomenon of  $\text{Sn}_7\text{Hg}$  by

preventing the formation of  $\text{Sn}_7\text{Hg}$  (gamma 2). However, even though the addition of Cu into amalgam alloys markedly improved the integrity of the dental amalgam, the  $\text{Cu}_6\text{Sn}_5$  (the eta prime phase of the copper amalgam system) is prone to corrosion according to the following reaction (Dental Materials, supra, at 293).



Furthermore, the copper which is known to leach out during corrosion of the high-copper amalgam systems has raised concern regarding the biocompatibility of these contemporary copper amalgams.

A more recent attempt to eliminate the well-known corrosive tendencies of the Ag-Sn and Ag-Sn-Cu amalgam systems is addressed in U.S. Pat. No. 4,181,757 which discloses a process for surface coating parent metals with a slurry (composed of a low-melting-temperature gold alloy powder, an inorganic oxide-dissolving flux, and organic liquid) and firing the slurry to produce a bonded, corrosion-resistant layer on the parent metal surface. The disclosed process is intended for use with dental prostheses and is not readily adaptable for in situ dental restorations such as fillings.

Additionally, recent evidence has raised new issues concerning the biocompatibility of currently used dental amalgams. Evidence has now been put forward that air expired by dental patients having dental restorations made from these amalgams, particularly after gum chewing, has a significantly higher mercury vapor content than that of amalgam free patients.

In an attempt to replace traditional dental amalgams with a more biocompatible intermetallic, experiments have been conducted on gallium alloys formed by triturating liquid gallium with a metal powder (Journal of Fukuoka Dental College, "Gallium Alloys for Dental Restorations," Takashi Horibe et al., 12(4): 198-204, 1986). However, the Journal of Dental Health has reported that gallium alloys supplied orally to rats and mice induce toxic responses in the test subjects (J. of Dental Health, "Study on Toxicity of a New Gallium Alloy for Dental Restorations," 37: 361-371, 1987). This raises questions about their safety as an alternative to traditional amalgams used in dental restorations.

## SUMMARY OF THE INVENTION

The invention pertains to an oxide-free metallic, alloy or intermetallic compound formed by coating a powder of at least one of either an elemental metallic, alloy or intermetallic compound with an oxide-replacing metal. The invention further pertains to a process for preparing an intermetallic alloy body by compacting the coated powder or a mixture of an elemental metallic powder and the coated powder without adding a liquid sintering agent to form an alloy body in situ. The compaction can be performed at temperatures below the melting points of the coated powder and a second elemental metallic powder and under sufficient pressure, to form a uniform alloy.

Intermetallic compounds and alloys produced in accordance with the present invention are substantially more biocompatible than existing commercial or experimental alloys.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is an x-ray diffraction spectrum for a compacted Ag-Sn intermetallic AGSN7.